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ABSTRACTS

COMPLEX DYNAMIC BEHAVIOUR IN THE RING REACTOR FOR METHANOL SYNTHESIS

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Complex Dynamic Behaviour in the Ring Reactor for Methanol Synthesis

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Forced unsteady-state catalytic reactors have received much attention in recent years, as concerns both industrial applications and scientific investigation and have been proposed both for endothermal and exothermal processes as well as for reversible and equilibrium reactions. Increased conversion, improved selectivity, reduced catalyst deactivation and autothermal behaviour are the main goals of such a mode of reactor operation.

The reactor network, or *ring reactor*, which consists of a closed sequence of two or more catalytic fixed bed reactors, with periodical variation of the feed position, is a way to obtain a sustained dynamic behaviour and to create a closed cycle, which prevents the heat front from leaving the system. This configuration has been recently investigated for the combustion of lean VOC mixtures. The autothermal behaviour, with a nearly uniform catalyst exploitation, due to the constant flow direction, is one of the main advantages of the network. It however presents a small range of switching times t_c which allow to reach and maintain a pseudo-steady state of operation. Previous works suggest that the reactors network can be a suitable alternative to reverse-flow operation, since it can reduce the emissions of unburned gas immediately after the change of the feed position [1]. Another advantage of this configuration is the possibility to achieve an optimal temperature distribution, which makes possible the creation of favourable thermodynamic conditions for exothermic equilibrium-limited reactions.

The methanol synthesis has been simulated with this configuration, proving that in

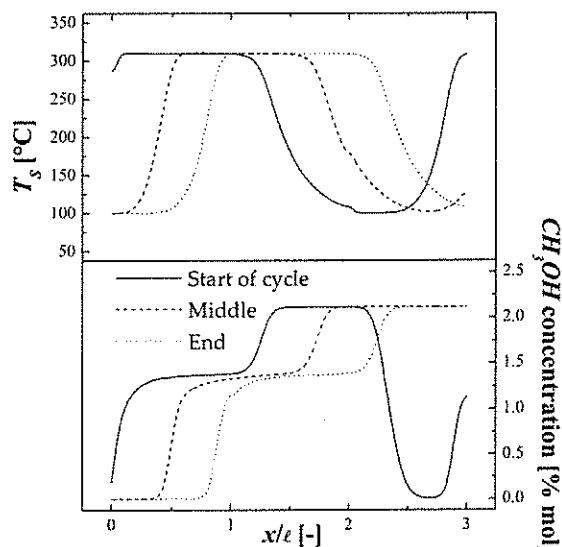


Figure 1. Methanol concentration and solid temperature profiles of the catalyst at the beginning, in the middle and at the end of the cycle after cyclic steady state has been reached; $t_c = 170$ s. $T_m = 100^\circ\text{C}$.

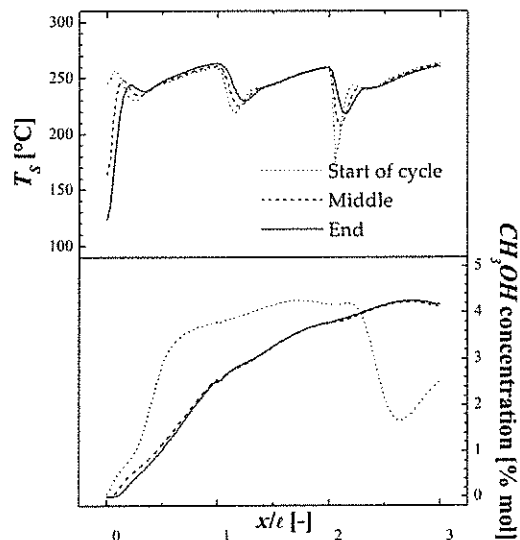


Figure 2. Methanol concentration and solid temperature profiles of the catalyst at the beginning, in the middle and at the end of the cycle after cyclic steady state has been reached; $t_c = 16$ s. $T_m = 100^\circ\text{C}$.

certain conditions the yield and selectivity of the process can be higher than in the reverse-flow operation and in the traditional multi-bed reactors with inter-refrigeration (the average carbon conversion is 58% against 30-40% of other devices). This is due to the temperature profile that can be obtained in the catalytic bed (Figure 1): the temperature decrease in the last bed of the sequence shifts methanol equilibrium concentration towards higher values. Nevertheless, final conversion in the network can be higher because a high temperature tail of the heat wave, which can re-ignite the partially converted gases coming from the first two beds at a concentration below the equilibrium value, thus giving an extra push in the

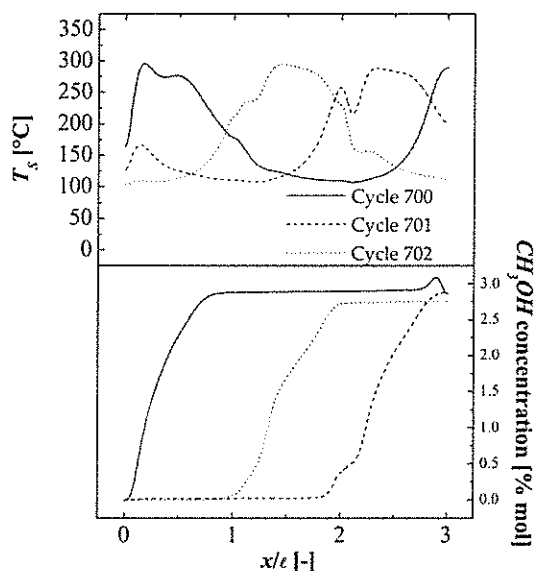


Figure 3. Temperature and methanol concentration profiles in the reactor corresponding to cycles n. 700, 701, 702; $t_c = 20$ s.

reached. In this case the profiles in a generic cycle are different from the others since the material and energy balances have a solution of period $47t_c$.

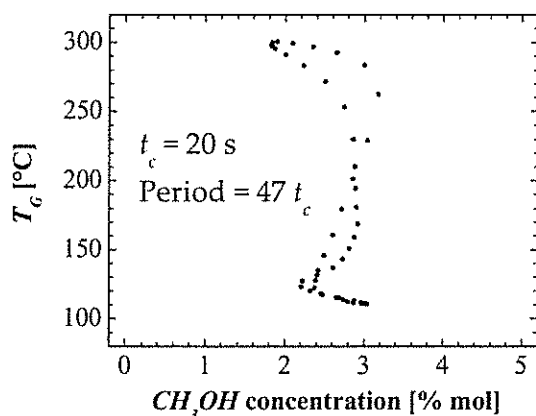


Figure 4. Correlation between methanol concentration and outlet gas temperature for $t = 1/2 t_c$.

conversion. In the case of optimal conversion ($T_{in} = 130^\circ\text{C}$, $t_c = 40$ s) the temperature profile approximates the curve corresponding to the maximum methanol generation. Better performances can be obtained reducing t_c . The result of the switching strategy is analogous to an intermediate cooling, which further increases the conversion, as it is shown in Figure 2. An additional increase of t_c finally leads to extinction of the reaction, as auto-thermal behaviour is not maintained for a wide intermediate range of cycle times. A narrow range also exists, close to the zone of maximum conversion, in which complex behaviours (cyclic steady-state of high periodicity) are observed. The temperature and concentration profiles for a switching time of 20 s, are shown in Figure 3. The three curves refer to the cycles 700, 701 and 702, when the pseudo steady-state conditions have been

The maps of Figure 4 show that for a certain instant during a cycle the reactor status is no longer defined by a unique couple of values (outlet gas temperature and methanol conversion), but 47 couples are possible, each one corresponding to one of the 47 cycles which form the operation period. If we change t_c also the period of the solution changes. As we move away from the stability operation range the period of the solution increases up to a value of $332t_c$ which corresponds to the upper limit of this high periodicity range.